MAGNETORESISTENCIA TUNEL EN JUNTURAS CON MANGANITAS

TUNNELING MAGNETORESISTANCE IN MANGANITE-BASED JUNCTIONS

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Resumen

Este artículo revisa la magnetoresistencia tunel (MRT) de junturas magnéticas, particularmente de aquellas construidas usando manganitas. Son presentados el origen de este tipo de tunelamiento y su dependencia con el material, el método de crecimiento de la juntura y la temperatura. El artículo es complementado con la discusión de algunos resultados sobre la influencia de la *medio-metalicidad* y de las intercaras en la MRT.

Palabras claves: manganitas, junturas magnéticas tipo túnel, magnetoresistencia túnel, half-metallicity

Abstract

This paper reviews the tunnelling magnetoresistance (TMR) of magnetic tunnelling junctions, particulary of those made of manganites. The origin of this type of tunnelling and its dependence on the material, the growing method and the temperature are presented. Besides, some results on the influence of the half-metallicity and of the interfaces on the TMR are discussed.

Keywords: manganites, magnetic tunneling junctions, tunnelling magnetoresistance, half-metallicity

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Introduction

The *spintronics*, or spin-dependent transport, meaning the use of the spin of the carriers for determining the direction and intensity of currents, has been widely applied since the discovery of the giant-magnetoresistance (GMR). In fact, the GMR has been used for magnetic information storage [1], as in read heads of hard-disc drivers.

Another possible application of the GMR is the fabrication of nonvolatile magnetic memories. However, for this purpose a different phenomena can be used, namely the spin-polarized tunnelling, discovered in the 70' by Julliere [2] who found it at low temperatures. In fact, he reported a variation in conductance of 14% at 4.2 K in Fe/Ge/Co devices by sweeping the external magnetic field. Thus, most transport properties are studied in magnetic tunnelling junctions (MTJ) made of two ferromagnetic electrodes separated by an insulating layer. This layer decouples magnetically the electrodes and the conductance of the device is then determined by the tunnelling due to the spin of the carriers. This is the so called tunnelling magnetoresistance. In MTJ's, the drop of the resistance is due to the change from the antiparallel configuration of the magnetization of the ferromagnetic electrodes to the parallel one by an external magnetic field.

Improving the TMR by different means is one of the main goals in spintronics. One possibility is the choice of the materials of which the MTJ is fabricated. The manganites, with a general formula $(R^{3+}_{1-x}A^{2+}_{2})MnO_3$ where R is a rare-earth element and A is an alkaline-earth, seem very promising to increase the TMR. In fact, junctions made of $La_{1-x}Sr_xMnO_3$ (LSMO) and $SrTiO_3$ (STO), (LSMO/STO/LSMO) revealed an extraordinary large TMR at low temperature with a resistance ratio of R_{high}/R_{low}=5-20 at 4.2K [3]. In addition, the LSMO optimally doped has a high Curie temperature ($\sim$ 370K), well above the room temperature and shows *half-metallicity*. This property can lead to an extremely high TMR.

However, one of the difficulties in the application of the manganites for spintronic devices is the lack of understanding of the role that interfaces play in MTJ. It is known that the TMR of

these junctions drastically decreases with temperature making difficult possible engineering applications. This behaviour seems to be strongly related to the interface between the manganite and the insulating barrier and several ways have been explored to understand why and how the properties of the interfaces determine the TMR.

This paper is built in the following way. First, the physics of the ferromagnetism in manganites is explained, then the phenomenon of tunnelling magnetoresistance is reviewed in different types of devices, particulary those made of manganites. Finally the half-metallicity of the manganites and its relation to the TMR is briefly discussed.

Crystal structure and double exchange mechanism in manganites

Manganites crystallize in the perovskite structure ABO_3, for example, the antiferromagnetic $LaMnO_3$ (fig. 1). It has practically a cubic structure with distortions of the lattice that result from stretching or tilting of the oxygen octahedra that surround the Mn ions. The replacement of some trivalent cations of La with divalent Sr cations gives rise to the presence of mixed 3+/4+ valence of Mn, as in the case of LSMO [4]. The discovery of metallic conductance and of ferromagnetism in doped manganites triggered the proposal by Zener of the so called double-exchange mechanism to explain these properties of the hole-doped manganites [3]. According to this model the localized 3d states of the Mn ions show a ferromagnetic interaction due to spin-polarized electrons: two electrons simultaneously are transfered, one from the Mn3+ ion to the O2p orbit and one from this orbital to the d orbit of Mn4+ [3].

Another important effect that takes place in manganites is the Jahn Teller distortion. It lifts the degeneracy of the e_g level with a splitting energy of around 1-1.5eV. As a result of this effect, in average all O_6 octahedra are distorted. These deformations can be long-range ordered or orbital order (OO). This orbital order, in manganites, comes usually together with superstructures of localized charge carriers or charge order(CO) [3]. The CO opposes the

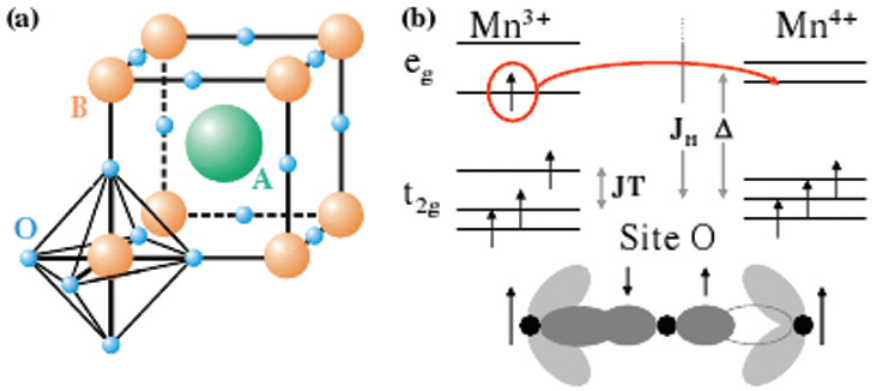

FIGURE 1. a) Crystal structure of the manganites b) The double-exchange mechanism between the σ Mn orbitals and the p oxygen orbitals. The degeneracy of the e_g and top t_{2g} levels of the Mn3+ is lifted by the Jahn Teller distortion of the oxygen octahedra [4]

double-exchange interaction.

The ground state of the doped manganites $(R_{1-x}A_x)MnO_3$ depends on a) the number x of doped carriers, b) the average ionic radii on La and Mn lattice sites and c)the scatter of ionic radii on La sites. The phase diagram dependence on the doping shows, in general, a metallic ferromagnetic region around $x = 0.33$ that is the largest in the La manganite doped with Sr, while it is absent for the Pr-Ca compounds.

Around $x = 0.5$ the manganites become antiferromagnetic and insulating and show CO. The same is true for the ground state at low doping. At high temperatures, manganites become insulating and paramagnetic. Among these three regions exist more complicated states that are still under research.

In the case of trilayers, as the LSMO/STO/LSMO, it has been found that the transport properties strongly depend on the interfaces. Thus the ferromagnetic coupling between the LSMO interface and the STO gets stronger in the case of electron doping, meaning the interface is a stacking of atomic planes MnO_2 /$La_{2/3}Sr_{1/3}O$/TiO_2, so called TiO_2 interface. Otherwise, the interface can be hole doping, like in the MnO_2/SrO sucession of planes or SrO interface. In the latter case, the ferromagentic cou-

pling is weakened [4].

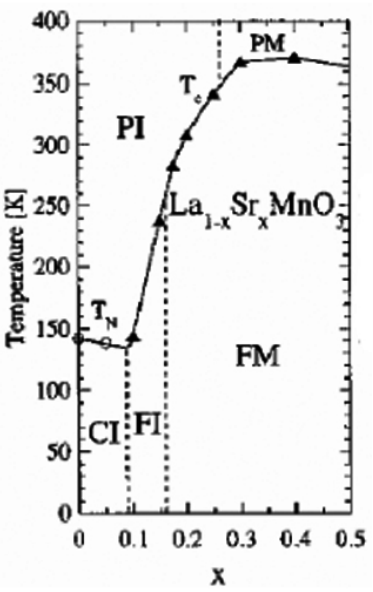

FIGURE 2. The phase diagram of the LSMO compound on dependence of the doping [3]

When designing devices based on manganites it is important to take into account the influence of the exchange interaction at the interface between the ferromagnetic and the antiferromagnetic layers. Kobrinskii *et .al* [5] studied the relationship between the interchange field (related to the shift of the hysteresis curve) and the thickness of the antiferomagnetic layer in the LCMO system. They found that the minimum thickness at which there is not shift of the hysteresis curve has a value of 50-100Å. Besides, as the AF layer thickness is increased, the shifting of the hysteresis goes to a constant value (saturation). Similar phenomema can take place in devices based on LSMO. In fact, the work by P. K. Muduli and R. C. Budhani [6] show that a AF layer with a 55% doping of Sr allows to generate a significant effect of exchange bias in a F layer (33% doping). These authors relate this behaviour to the magnetoresistance of a MTJ where the second electrode is Co and the insulating layer is $SrTiO_3$. However, the values of the TMR studied by Muduli and Budhani are low even if one takes as reference the values for transitions metals that have shown a TMR of 40% at helium temperature and 29% at room temperature [7].

Spin polarization and tunnelling magnetoresistance

When Julliere [2] discovered the TMR, he proposed that it was mainly govern by the relative magnetization orientation of the ferro-

magnetic electrodes. In a first approximation the spin polarization (SP) P of the junction can be modeled as due exclusively to the density of states (DOS) at the Fermi level, of the mayority-spin electrons $\rho^{\uparrow}$ and that of the minority-spin electrons, $\rho^{\downarrow}$, in the ferromagnetic electrode:

$$P = \frac{\rho^{\uparrow} - \rho^{\downarrow}}{\rho^{\uparrow} + \rho^{\downarrow}} \tag{1}$$

Thus, the conduction in ferromagnets can be explained by the existence of two subbands where the spin-up subband corresponds to those electrons with spin parallel to the magnetization, and those with spin-down are in the minority-spin subband. Besides, these two bands are shifted respect to each other. If there is not spin-flipping the two subbands do not mix. However, this simplest model does not take into account that the conductance depends not only on the number of electrons with a given spin, but also on their tunnelling probability. The latter depends on the band structure of the ferromagnet. This understanding was reached through measurements of the conductance of MTJ in which one of the electrodes was a superconductor. However, when using a junction made of FM/I/FM in which the spin detection is due to the exchange-split states of one of the ferromagnets, the relative magnetization orientation gives rise to the TMR. Then the TMR is defined in terms of the difference of the resistances of the parallel, R_p, and antiparallel, R_{ap}, magnetizations [8], [9]:

$$TMR = \frac{R_{ap} - R_p}{R_p} = \frac{2P_1P_2}{1 - P_1P_2} \tag{2}$$

where P_1 and P_2 are the polarizations of each electrode.

This is the so called optimistic definition, while the pessimistic one is calculated respect to the R_{ap}.

According to (fig. 2) the TMR could depend only on the relative orientation of the spins in the two ferromagnetic electrodes. However, recent experiments have shown that the magnitude of the TMR depends strongly on the intrinsic characteristics of the MTJ like the interface metal/insulator [8]. For example, the first results for the SP of permalloy was about 32% while after improving the fabrication techniques, the TMR increased up to 48%. Moreover,

de Teresa *et al.* [10] found, using LSMO as one electrode in a MTJ, that depending on the insulating barrier the TMR could be positive or negative. In fact, when working with $Co/Al_2O_3/LSMO$, the TMR was positive while when using $SrTiO_3$ as barrier, the TMR was negative (fig. 3). This was explained due to the bonding states at the barrier interface. Thus the TMR has to be thought in terms

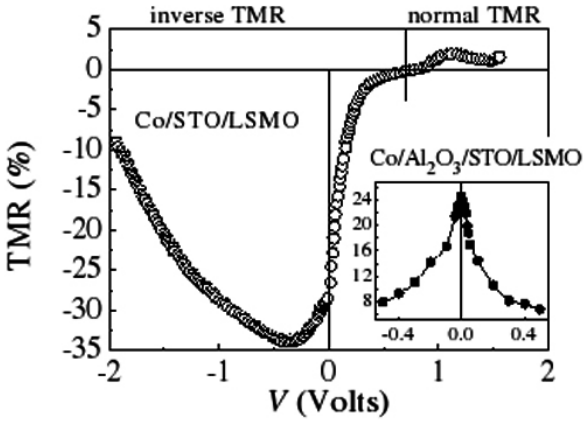

FIGURE 3. TMR of a $Co/SrTiO_3/LSMO$ junction. As seen up to 0.8V the TMR ratio is negative, indicating that below this value the $Co/SrTiO_3$ spin polarization must be negative. The inset shows the TMR for a $Co/Al_2O_3/STO/LSMO$ junction [10]

not just of the SP of the magnetic electrodes but rather on terms of their interfaces.

Another research on this topic is the one by Viret *et al.* [11], who fabricated trilayers of LSMO with three different barriers. The LSMO layers had a thickness of 35nm and 25nm, while the insulating layer was just 3nm thick and with a junction area of $6\times6\mu$m. Three different compounds were studied as barriers, namely $PrBa_2(CuGa)_3O_7$, CeO_2 and $SrTiO_3$ finding with this last barrier a SP of 83% at low temperatures. This high polarization can be understood as a proof of few hybridization at the Fermi level of the Mn_{3d} levels with the O_{2p} ones, so that the level has mainly a 3d character [11]. On the other hand, it was found that the TMR decreases with the temperature starting at a temperature

of 190K. For comparison, in fig. 4 the temperature dependence of the resistance of the junction and of the bottom electrode are shown. The reduction of the resistance of the MTJ is typical of

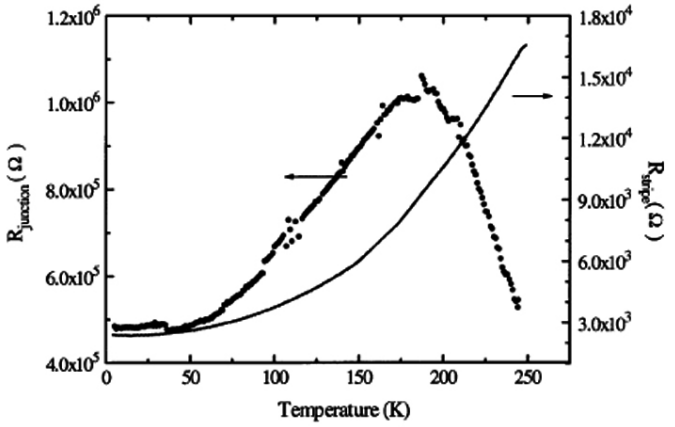

FIGURE 4. Temperature dependence of the resistance of the MTJ showing a maximum at 190K and of the bottom electrode that shows the behaviour of a good $La_{0.7}Sr_{0.3}MnO_3$ [11]

the underdoped manganites,thus this behaviour could be related to a lack of oxygen at the F/I interface, leading to spin-flipping. Researches on the origin of the dead layer have been carried out on LSMO/STO superlattices and LSMO/STO/LSMO trilayers fabricated by pulsed-laser deposition [12]. The main conclusion of the work by Ogimoto *et al.* [12] is that the reduction of the TMR of MTJ based on LSMO at high temperatures for different doping depends on spin canting at the interface. When using $x = 0.3$, a compromise between high T_c and lower spin canting seems to be reached as can be inferred from the TMR resistance that survives up to 320K. Besides, the performance of the MTJ depends on the fabrication technique which affects the interface quality, stoichiometry and the TMR. The MR measured by Ogimoto *et al.* at 5K was only of 12%. This is one reason why the molecular beam epitaxy (MBE) technique is one of the most used, since it makes easier to control the growing and arquitecture the interfaces. One of the earliest research in this direction by O' Donell *et al.* [14], controlling

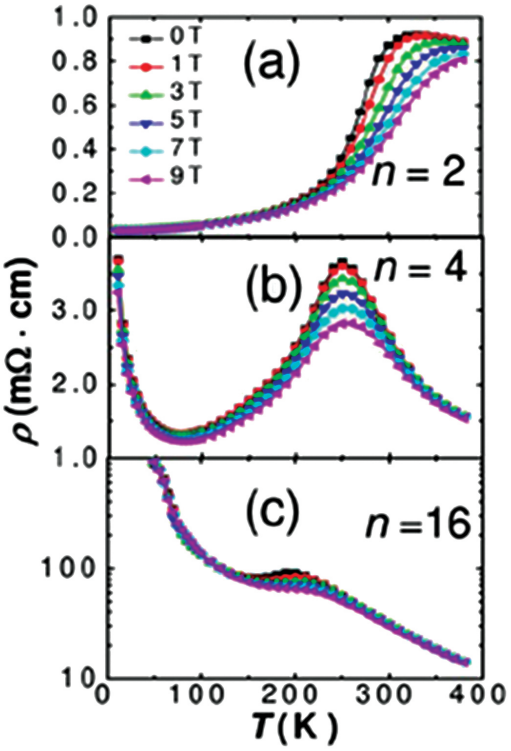

FIGURE 5. Resistivity vs. Temperature for superlattices $[(SrMnO_3)_n/(LaMnO_3)_{2n}]_m$ with different n and under different applied in plane field [13]

the layer-by-layer growth with RHEED, showed that those trilayers with some degree of disorder also had a lower MR that faster vanished with temperature. Some investigations have been carried out on superlattices made of antiferromagnetic layers but that exhibit, depending on the number of layers, ferromagnetic behaviour. In fact, in $[(SrMnO_3)_n/(LaMnO_3)_{2n}]_m$ superlattices grown by MBE, it has been found [13] that for n$\leq$ 2 they show the same behaviour as the optimal-doped oxide manganite $L_{0.67}S_{0.33}MO_3$ and when increasing n the superlattice becomes insulating, but more conductive than the separate components (fig. 5).

The former property could be explained due to the mixing of

the Mn ions valance as explained by the exchange model, but in this superlattices it happens at the interfaces, since Mn is 4+ at the SMO layers and 3+ at the LMO layers. As the number of layers (n) increases, this interaction is smashed out. Surprisingly, all samples even with big n showed a large magnetization, a feature that was explained as due to an excess of oxygen in the LMO layers.

Summarizing up, a complete model of the TMR should take into account:

1. The band structure of the electrodes.
2. The tunnel barrier characteristics, as the interfaces.
3. The possible spin-flip processes.

The half-metallicity of manganites

When using as electrodes half-metals it is teoretically possible to reach an infinite TMR, since one should replace P_1=$P_2 \sim 1$ in eq.2 . Basically, half-metals are those materials that appear to have charge carriers with only one spin direction at the fermi level, E_F. The 100% SP brings a unique opportunity of developing a whole new electronic technology. The new devices could be smaller and possess intrinsic memories. For example, logic gates could be fabricated based on half-metals. These devices could keep their function even after cutting the power off. Moreover, they could be reset to serve other functions by reversing the magnetization of some elements since 100% polarizations permits true on/off operation [1]. This opens the possibility of using a standardized reprogrammable logic chip as a universal microprocessor. However, one of the difficulties in the application of the manganites for spintronic devices is, as mentioned in the previous section, the lack of understanding of the role that interfaces play in MTJ.

Few compounds have been predicted to show half metallicity, they are CrO_2, Fe_3O_4, mixed-valence manganites and Heusler alloys [15]. It is interesting to notice that many compounds that content Mn as NiMnSb, $La_{0.7}Sr_{0.3}MnO_3$, $Tl_2Mn_2O_7$ and Mn doped

semiconductors (e.g. (Ga,Mn)As) have a high SP, while the element Mn is antiferromagnetic [3]. The TMR of MTJ's based on LSMO rapidly decreases with the temperature. Among the possible explanations for this behaviour are:

1. The formation of a dead, paramagnetic, layer close to the interfaces. This layer reduces drastically the SP by spin-flipping of the carriers going through the barrier [14].

2. The existence of another tunnelling channel that works at high temperatures and it is not spin polarized, mainly through defects on the interfaces [14].

3. The reduced oxygen content of the LSMO/STO interfaces [11]

The half-metallicity of the manganites has been under discussion for more than a decade. One reason is because it is extremly difficult to get experimentally a 100% polarization. Since the magnetotransport of the devices made of these materials rely on this property, some direct experiments have been carried out in order to establish it. For example, the differential conductance as a function of the temperature and the applied voltage has been measured by tunnelling spectroscopy of thin films of several manganites, among them, LSMO with different doping [16]. Weia *et al.* [16] found that the energy gap of the minority carriers is smaller for the Sr doped manganite, LSMO, than the Ca doped or LCMO. This seems to be in agreement with a higher hybradization of the p-d orbitals due to a bigger lattice distortion in the LCMO than in the LSMO. Spin-resolved photoemission measurements have also shed light on this question. According to this technique the Heusler alloy NiMnSb shows only a 50% SP while for the ferromagnetic oxide CrO_2, the SP is 100% but only at 2eV binding energy and not at the E_F level. These results could be due to the fact that the spin-resolved photoemission measurements are surface sensitive and these two materials show a non stoichiometric composition precisely on the surface and this is the reason for the low SP measured by this technique. Park *et al.* [17] studied thin films of the oxide manganite $La_{0.7}Sr_{0.3}MnO_3$ by this technique and were able to prove that (i) far below T_C the SP is 100% (ii) there is a transition from the metal-ferromagnetic

state to a pseudogap state when approaching T_C. This transition

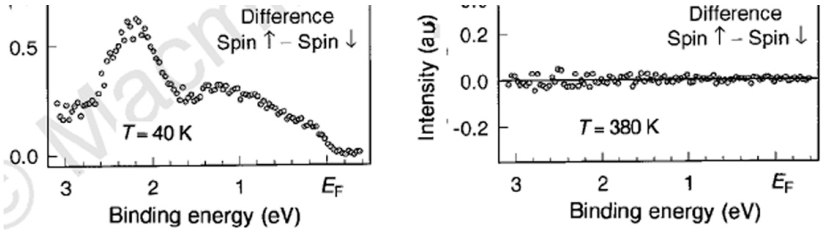

FIGURE 6. The difference of spin-resolved photoemission spectra between the mayority and minority carriers at two temperatures well below Tc, 40K, and well above Tc, 380K of $La_{0.7}Sr_{0.3}MnO_3$ thin film [17]

can be explained by the change of the interaction of the mixed valence ions. Below T_C, the Mn_{3d} electrons are strongly polarized by the double-exchange interaction. When increasing the temperature the spin anisotropy progressively dissapears and the hoping electron energy decreases and together with the Jahn-Teller distortion both lead to the absence of states at the E_F. On the other hand, the O_{2p} states are not polarized, meaning there are mayority and minority carriers and for this reason at low temperatures the minority states show a gap that is between the O_{2p} minority states and the unocuppied Mn_{3d} minority states.

In any case, the LSMO-based junctions have possibly shown the largest TMR [3]. A TMR of 1800% [15] was found in a LSMO(35)/STO(2.8)/LSMO(10) junction (the numbers in parenthesis indicate the thickness of each layer in nm). According to Bowen *et al.* [15] such a high TMR is reached for small area junctions. The devices of 5,6 × 5,6 μm ^{2} area showed a TMR of 1850% at 4K and excitation voltage V_{DC} =1mV. For smaller junctions 2× 6 μm ^{2} the TMR dissapears only at high temperatures as 280K (see fig. 7) To improve the coercive field of the top LSMO layer, Co and CoO layers were grown on the top of the junction. Unfortunately, they haven't report such high TMR on more samples. This research group claims [4], that polarizations of 95% and of 99% can be obtained depending on the bias , being smaller at low bias and larger in the opposite case. This could be explained because at low bias, spin waves are excited. A more recent paper by Werner *et*

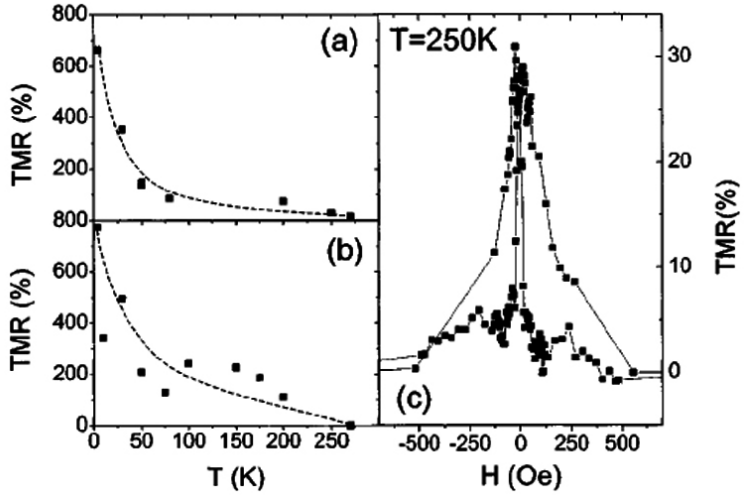

FIGURE 7. Temperature dependence of TMR for two MTJ with different area a) $2 \times 6\mu m^2$ and b) $1.4 \times 4.2\mu m^2$ and c) R(H) at 250K that shows a TMR of 50% [15]

al. [18] reported a TMR as high as 1900%. Besides, it was found that the TMR has a four-fold symmetry. This shows that uniaxial anisotropy is not necessary for large TMR [18]

Moreover, the LSMO/STO is considered as the best system to study the half-metallicity theoretically as well as experimentally. Garcia *et al.*[19] investigated the SP of interfaces, determining it from TMR measurements of junctions LSMO/insulator/LSMO using as barriers $SrTiO_3$, TiO_2, $LaAlO_3$. They found that for all barriers the behaviour of the magnetization with the temperature is similar to that of the bulk magnetization but shows a lower Curie temperature. In contrast, when comparing this behaviour with that of a free surface, the result is quite different. The reduced magnetization of the free surface, measured by spin-polarized photoemission spectroscopy, can be understood as a proof of the role of the oxygen octahedra in the double-exchange mechanism and thus, in the ferromagnetism of interfaces and free surfaces. In the latter, the octahedra are distorted stronger or even not complete, the SP decreases faster with the temperature than the SP of interfaces.

Moon *et al.* [20] studied magnetic tunnelling junctions of

$La_{0.7}Ca_{0.3}MnO_3$ (LCMO) with $NdGaO_3$ as barrier. The trilayers were grown by pulsed-laser deposition and showed between 120 K and 300K a non-tunnelling conductance due maybe to defects of the barrier. The TMR gets reduced by 40% at 100K and completely dissapears at 150 K. This reduction can be explained by a percolative process of phase separation at the interface. Thus, a better lattice matching between the electrodes and the barrier can keep a high TMR. Giving the low TMR at high temperatures, some theoretical

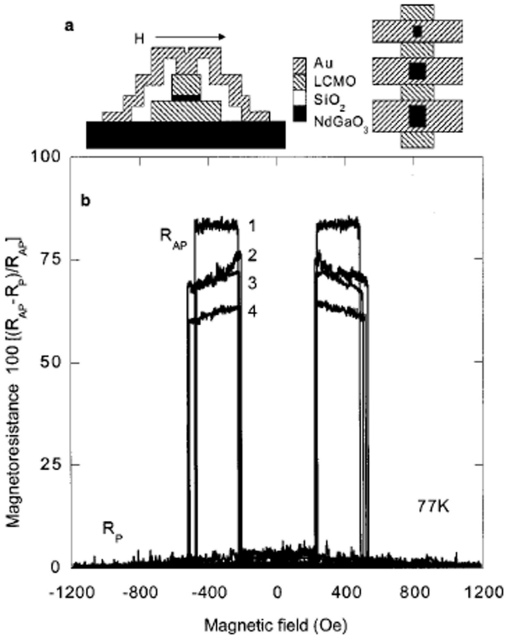

FIGURE 8. a) Cross section of the MTJ made of LCMO/NGO/LCMO b) TMR at 77K of junctions of different areas 1: $6 \times 6\mu m^2$, 2:$8 \times 8\mu m^2$, 3:$8 \times 16\mu m^2$, 4:$20 \times 30\mu m^2$ [20]

studies on manganites have proposed to use as barrier $LaMnO_3$ [21]. In this work the magnetic coupling of the LSMO electrodes depend on the number of LMO layers, for an even number of them, the magnetization of the electrodes will be antiparallel and the opposite for an odd number. However, this coupling is weak but still a large TMR is expected at low temperatures. Several approximations are made in this theoretical analysis. Among them, no dead

layer was modeled, no lattice ralaxation between LMO/LSMO and not complete anysotropy was taken into account.

Besides the trilayer junctions, other devices can be fabricated from LSMO to make use of the half-metalicity. Among these devices there are ramp-edge and step-edge junctions. Wanga *et al.* [22] fabricated and studied step-edge junctions of LSMO. They found a butterfly like behaviour of the magnetic resistance of the junctions depending on the direction of the applied magnetic field. The direction of the field is measured between the surface of the sample and the field as shown in fig.9. The anisotropy of the TMR might be due

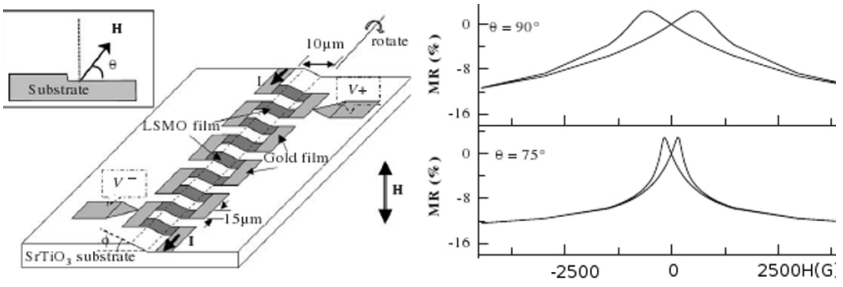

FIGURE 9. *left*: Scheme of the step-edge junction. *right*:The butterfly effect of the MR of the step-edge junction at 100K for two different angles of incidence of the magnetic field [22]

to the induced magnetization in the steps of the grain boundaries. Also, there have been already attemps [23] of combining LSMO with Si looking for applications for the semiconducting technology. The $La_{0.7}Sr_{0.3}MnO_3$ polycrystalline films grown on Si covered by naturally amorphous SiO_x show strong x-ray magnetic circular dichroism (XMCD) signal that implies a high SP at the surface.

Cespedes *et al.* [24] have studied the transport properties of nanoconstrictions of two materials that show a high SP at room temperature, namely Fe_3O_4 and LSMO. The restrictions have a width between 20-50nm of the order of the domain wall. The IV characteristics (fig.10) can be fitted to a third power order of the voltage (i.e. the same type of dependence as for tunnel junctions) In this work not all nanoconstrictions show magnetoresistance. It is proposed that the absence of it is due to the absence of a domain wall at the constriction, since it will in fact affect the electron

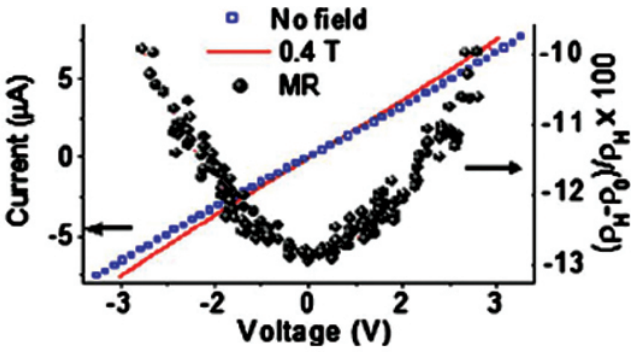

FIGURE 10. Current and magnetoresistance dependence on voltage of a LSMO nanoconstriction [24]

current through the structure. This seems also to be in agreement with the decreasing of the coefficient corresponding to the nonlinear term of the IV fitting. In fact, they found that this coefficient gets smaller when a field is applied, meaning probably the domain wall is shifted letting the carriers to move from one side of the constriction to the opposite. Before the wall is shifted, the behaviour is quite different, the nonlinear behaviour is significant as the transport is maybe due to tunnelling through the domain wall.

Conclusion

The study of MTJ's, particulary those based on manganites, has achieved interesting results, like high TMR values [18]. However, the TMR is very sensitive to temperature and to interface characteristics. These two factors can drastically reduce the magnetoresistance of the tunnelling junction. This could be due to some mixing of the, ideally, spin-independent conduction channels. Some spin scattering on the interfaces can take place as well as spin-flipping due, for example, to roughness of the interfaces and to thermal activation.

More realistic models of the tunnelling in such devices are needed together with the improvement of the growing techniques, especially the growing of the interfaces.

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